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Anomalous enhancement in the optical scattered radiation in magnetite base ferrofluid

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Abstract

Analysis and measurements of optical transmission of laser beam through ferrofluid have shown anomalous optical behavior of kerosene-based ferrofluid under the presence or absence of magnetic field. The optical transmission is polarization dependent and the observed pattern is attributed to dipole scattering. The spectral characteristic of ferrofluid indicates the dipole scattering and superparamagnetic behavior of the particles. The anomaly in the system is correlated with the magnetic, size distribution and other spectral measurements. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

Particle size distribution and number densities are important aspects in a stable ferrofluid. The number density of the particles in stable ferrofluid is usually very high, due to which they experience short-range attractive and repulsive forces. Magnetic particles in a ferrofluid with their large magnetic dipole moment have many interesting properties spanning from superparamagnetism to ferromagnetism. Investigations on aggregates, turbidity and spatial ordering in magnetic fluids have been made by using magneto-optical methods [1]. The functions of various ferrofluid devices like optical modulator and birefringence viscometer, etc. are governed by the orientation distribution of particles. The scattering by the individual particle in stable ferrofluid determines the optical properties of the fluid as a whole. Martinet et al. [2] have explored the possibility of using a magnetic fluid as an optical shutter. There are several theories which explain the magneto-optical effect like particle orientation, chain formation, rotation, etc. The domain magnetization and concentration are very important in determining the response of the fluid in the applied magnetic field. The present study aims at

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finding the changes in the magnetically induced scattered light intensity in kerosene-based magnetic fluid, optical reflectance of thin film grown under the presence or absence of magnetic field and also their magnetic measurements.

2. Experimental details

A kerosene-based magnetic fluid with Fe₃O₄ ferrimagnetic particle obtained from Tohoku University, Sendai, Japan, was used for the present investigation. The particle size distribution is in the range $\sim 10-150$ Å. The saturation magnetization of the fluid was about ~400 G, density 1.359 g/cm³. A light beam of He–Neon laser of 2mW was used to study the magnetically induced scattered intensity. A laser light passes through a polarizer and a quarter waveplate, which together enable an adjustment such that two eigenmodes are equally excited with varying magnetic field (~ 1000 G) at the input of the specially made ferrofluid sample cell $(20 \times 10 \text{ mm}^2 \text{ diameter})$. Various sets of measurements were also carried out by diluting the ferrofluid concentration at 1:2:3 ratio. The magnetic field was placed normal to the beam of light to observe the magnetically induced scattered light intensity from the ferrofluid



Fig. 1. A schematic diagram of optical experimental setup for magnetically induced light scattering measurement.

sample. The output of the induced beam was then recorded by using a spectrum analyzer. The spectrum analyzer constituted of double grating monochromator attached to thermoelectrically cooled photomultiplier. The experimental setup is shown in Fig. 1. For reflection and magnetic measurements two sets of thin films on a glass substrate were prepared using spinning technique [3]. The films were grown under the influence of magnetic field (~ 1000 G, film a) and without magnetic field (film b). These films were characterized to understand if there was any change in the physical properties and their behavior. For crystalline phase analysis X-ray diffraction studies were carried using Siemen D-500 powder X-ray diffractometer, Cu K_{α} radiation. Goino reflectometer was used to measure the reflectance of the films and the magnetization measurements were performed by using DMS 880 vibrating magnetometer.

3. Results and discussions

3.1. Magneto-optic effect in ferrofluid

In the presence of magnetic field many ferrofluids exhibit birefringence, dichroism and Faraday rotation [4-6] in the visible region of the electromagnetic spectrum. Formation of the chain like cluster of ferrofluid particles under various physical conditions (like concentration, external field, temperature, etc.) have been studied extensively [7–9]. The result of our study carried out for the magneto-optical effect of 50% dilution kerosene-based ferrofluid at magnetic field (1000 G) is depicted in Fig 2. The induced scattered intensity of light through the ferrofluid cell shows enhancement in intensity in the presence of magnetic field as compared to that in the absence of magnetic field. Similar observation have been seen at different magnetic field and density. Further increase in the magnetic field does not show any increase in the induced light intensity. The variation in the intensity of induced light was found to vary exponentially on varying the applied magnetic field. This variation may be attributed to the percentage variation of superparamagnetic (SP) particle on application of magnetic field. The non-SP

particles influence the SP particle in their vicinity and hinder the movement of the particles.

3.2. Optical reflectance

Almost all the ferrofluid particle possess spinel structure. This is a cubic closed packed cage of oxygen ions with the metallic ions occupying the tetrahedral A and octahedral B interstitial site. The distance between two metal ions in octahedral B sites is smaller than the distance between metal ion at B site and another metal ion at tetrahedral A site. The electron hopping between B and A sites under normal condition will have less probability as compared to B–B hoping. The hopping process depends upon the separation between ions involved and the activation energy.

The optical reflectance study carried out for both the films at 25° incidence angle shows an increase in the reflectance by 15% for the film (a) as compared to the film (b). This may be due to charge transfer in orientation of domains and less electron hopping in the ferrofluid film. A comparative optical reflectance spectrum of the films is shown in Fig. 3. In the visible region spin flip transition $Fe^{3+} + Fe^{3+} \rightarrow Fe^{2+} + Fe^{4+}$ accounts for the spectral feature. The (e^-, h^+) pair thus generated are assumed to possess low mobility (high localized states). Because of the low absorptivity of the sample, the (e^{-}, h^{+}) pairs generated are distributed in the ordered chain like clustering of the ferrofluid particles and increasing the percent reflectivity. Another possibility arises due to the set array of absorbed surfactant carbon molecule in the film.

3.3. Magnetic measurements

For any ferromagnetic material, magnetic moment per unit volume or magnetization is a measure of its magnetic strength and the manner in which it varies in the applied field decides its utility. The results of studies carried out on magnetic moment versus applied magnetic field (7×10^3 Oe) at room temperature of the film (a) using VSM is shown in Fig 4. Film prepared under the influence of magnetic field shows higher magnetic moment 0.0140 emu as compared to the film prepared without field 0.0138 emu. This behavior could be related



Fig. 2. Induced scattered light intensity spectrum of ferrofluid sample.



Fig. 3. Optical reflectance spectrum of the films (a) and (b).

to the presence of variation in the particle size distribution. The particle in the SP size limit may be getting aligned in the direction of applied magnetic field and leading to larger magnetic moment. Ferrofluid as we know usually consists of the particle of SP size range. The SP behavior is observed when the thermal energy kT of the particles exceeds over the magnetic energy kV. The M-H curve and the magnetization characteristics suggest the SP size range of the particles in ferrofluid films.

4. Conclusion

The present study reveals that the majority of the particles are of SP size. These nano-scale particles



Fig. 4. Magnetization curve of film (a).

exhibit exotic feature as compared to their bulk. A study on the orientation of the domains under various physical parameters like concentration, external magnetic field and temperature may bring deeper insight in the association phenomena in a magnetic fluid. The film grown under the influence of magnetic field has shown the changes in the physical properties as compared to film without field and exhibit quite interesting results.

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